

Hyperfine Interactions in the Heavy Fermion CeMIn₅ Systems

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Abstract. The CeMIn₅ heavy fermion compounds have attracted enormous interest since their discovery six years ago. These materials exhibit a rich spectrum of unusual correlated electron behavior, and may be an ideal model for the high temperature superconductors. As many of these systems are either antiferromagnets, or lie close to an antiferromagnetic phase boundary, it is crucial to understand the behavior of the dynamic and static magnetism. Since neutron scattering is difficult in these materials, often the primary source of information about the magnetic fluctuations is Nuclear Magnetic Resonance (NMR). Therefore, it is crucial to have a detailed understanding of how the nuclear moments interact with conduction electrons and the local moments present in these systems. Here we present a detailed analysis of the hyperfine coupling based on anisotropic hyperfine coupling tensors between nuclear moments and local moments. Because the couplings are symmetric with respect to bond axes rather than crystal lattice directions, the nuclear sites can experience non-vanishing hyperfine fields even in high symmetry sites.

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1. Introduction to NMR in the CeMIn_5 Materials

The CeMIn_5 materials (also known as the 115's), with $M = \text{Co, Rh or Ir}$, exhibit a rich variety of phenomena as a result of strong electron-electron correlation [1]. These phenomena include unconventional d-wave superconductivity, local moment magnetism, coexistence of magnetism and superconductivity, and a possible Fulde-Ferrell-Larkin-Ovchinnikov superconducting phase [2, 3]. The superconducting transition temperature, T_c , of CeCoIn_5 is the highest of the Ce-based heavy fermion materials. Recently, an isostructural class of plutonium based 115's was synthesized, the PuMGa_5 series, with $M = \text{Co or Rh}$, that are superconductors with T_c 's of 18.5K, and 8.5K, respectively [4]. Since PuCoGa_5 is a d-wave superconductor like CeCoIn_5 , it has been argued that its ten-fold increase in T_c is due to an increase in the size of the relevant magnetic interaction energy, J , and thus the PuMGa_5 materials bridge the gap between two distinct sets of unconventional superconductors: the heavy fermion superconductors, with $T_c \sim 1 - 2\text{K}$, and the high temperature superconductors, with $T_c \sim 100\text{K}$ [5]. Consequently, one can reasonably argue that by investigating the role of magnetism and superconductivity in the CeMIn_5 materials, one can gain considerable insight in the the role of magnetism and superconductivity in the high- T_c materials and perhaps shed new light on the details of the pairing mechanism. These heavy fermion superconductors allow us to probe the physics over a much broader temperature range compared to the characteristic spin fluctuation energy, J (where J typically is on the order of 1500K for the cuprates and 10K for the heavy fermions). Furthermore, the CeMIn_5 materials are more easily handled in the laboratory, can be tuned with pressure rather than doping, and are extremely clean with small residual resistivities and large scattering lengths.

The magnetism in the CeMIn_5 system is clearly relevant at some level for the existence of the unconventional superconductivity and the non-Fermi liquid behaviors [6, 7, 8, 9]. The magnetism can be investigated experimentally by measuring the dynamical magnetic susceptibility experimentally, either with neutron scattering or NMR. Neutron scattering experiments often are limited because the ^{115}In strongly absorbs neutrons (cross section ~ 200 barns), which severely limits the resolution. On the other hand, NMR can provide important insight into the behavior of the dynamical susceptibility in these systems through measurements of the spin lattice relaxation rate, T_1^{-1} , and the Knight shift, K , of the In, Ga or M nuclei. Since the nuclear spins are coupled to the electron spin moments in the system, they are sensitive to the dynamics of these electron spins. In fact, T_1^{-1} , is sensitive to the imaginary part of the dynamical electron spin susceptibility, whereas K probes the static susceptibility. The details of how the nuclei couples to the electron spins, however, are important for interpreting the T_1^{-1} and K data. For example, if a nucleus is coupled to more than one electron spin, then the hyperfine field can vanish for particular spin configurations at certain nuclear sites, depending on the symmetry of the site.

In this manuscript we attempt to provide a complete analysis of the hyperfine couplings to the various NMR-active sites in this 115's. Since their discovery six years

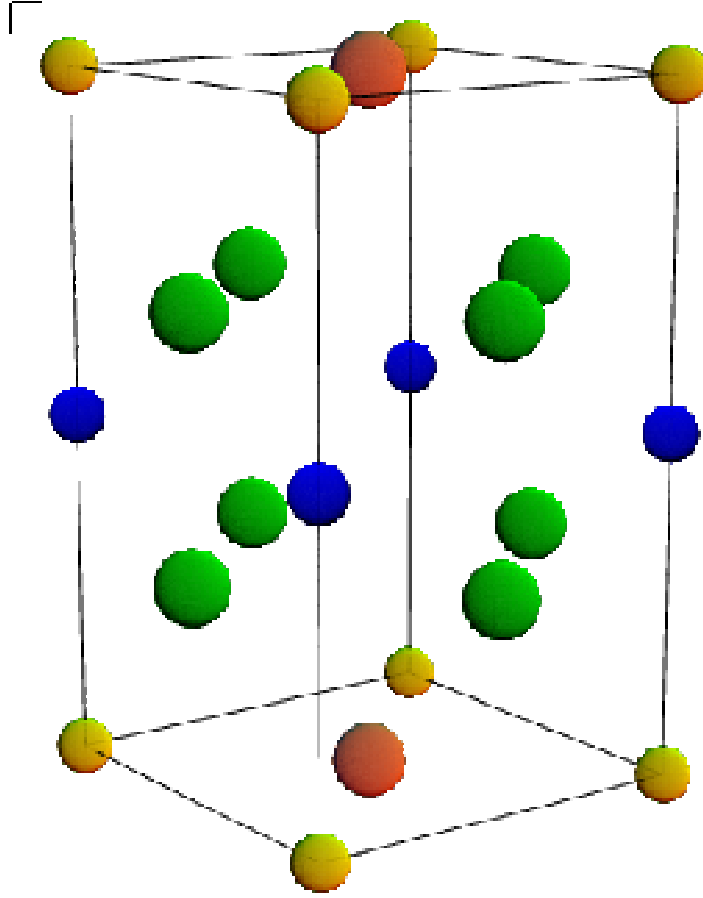


Figure 1. The structure of the $CeMIn_5$ materials. The Ce (Wykyoff position 1a) atoms are yellow, the In(1) (1c) atoms are orange, the In(2) (4i) are green, and the M atoms are blue (1b). Details of the structural parameters can be found in [10].

ago, several groups have reported Knight shift and spin lattice relaxation rate data in these materials. However, in order to make any connection between the NMR data and physically relevant quantities such as the dynamic or static susceptibility, one needs to understand the hyperfine interaction. Shortly after the first NMR experiments in the cuprates, Mila and Rice showed that the response of the Cu and O can be understood in a relatively simple single spin component model by elucidating the hyperfine interactions [12]. Their pioneering work led to significant insight into the cuprates as observed by NMR. The hyperfine scenario in the 115's, and the heavy fermion compounds in general, is more complex than that in the cuprates. There is strong evidence for two components of spin susceptibility, with different temperature dependences. Since the spin lattice relaxation is dominated by fluctuating hyperfine fields, these two components can contribute in unexpected ways to the temperature dependence. Consequently, one must approach any interpretation of T_1^{-1} data with an eye to these anomalous effects, and a complete analysis requires an intimate understanding of the hyperfine interactions.

2. Transferred Hyperfine Couplings

Most of our knowledge of the hyperfine interactions in the 115's comes from measurements of K and the static hyperfine field, H_{hyp} in the magnetically ordered state. In general, a nuclear spin $\hat{\mathbf{I}}$ is coupled to an electron spin $\hat{\mathbf{S}}$ via an interaction, A :

$$\hat{\mathcal{H}}_{\text{hyp}} = \gamma \hbar A \hat{\mathbf{I}} \cdot \hat{\mathbf{S}}, \quad (1)$$

where γ is the gyromagnetic ratio. In an external field H_0 , the electron spins are polarized via their uniform (i.e. $\mathbf{q}=0$) susceptibility: $M = g\mu_B \langle \hat{\mathbf{S}} \rangle = \chi H_0$. This leads to a shift, K , in the NMR resonance frequency:

$$\hat{\mathcal{H}} = \gamma \hbar \hat{\mathbf{I}} (1 + K) \cdot \mathbf{H}_0, \quad (2)$$

where $K = A\chi/g\mu_B$. If the susceptibility is strongly temperature dependent, then the Knight shift should be as well. By measuring $K(T)$ and $\chi(T)$ independently, and plotting K versus χ with T as an implicit parameter, one can extract the value of the hyperfine coupling constant, A .

In the CeMnIn_5 materials, χ is strongly temperature dependent with Curie-Weiss behavior. There are two sets of spins that potentially can give rise to this susceptibility: the conduction electron spins, $S_c = 1/2$, and the local moments of the Ce^{3+} 4f¹ electrons, S_f . Note that for the lanthanides, the spin-orbit interaction is several eV, so for experimentally realizable temperatures, the important quantum number is the total spin $J = L + S$, where $L = 3$ and $S = 1/2$ for Ce^{3+} . The degeneracy of the ground state 5/2 multiplet is further lifted by the crystal field interaction, which has tetragonal symmetry in the 115's, typically on the order of 50-100K, so that at low temperatures the ground state of the Ce^{3+} is a pseudospin doublet with an effective g value determined by the crystal field. For simplicity, we denote the 4f spin by S_f . The local moments and the conduction electrons also experience a Kondo interaction and an exchange interaction between different 4f spins that is driven by an RKKY or other similar mechanism. The hyperfine interaction couples the nuclei to these two sets of spins, but the coupling strength is several orders of magnitude smaller than the other interactions in the system. Therefore, the nuclei are sensitive probes of the magnetic behavior of the system, but they do not significantly perturb the intrinsic phenomena. Empirically, we find that for sufficiently large temperatures $T > T^*$, where T^* is material dependent but on the order of 50K, K is linearly proportional to χ . Since the dominant contribution to the susceptibility is from the local moments, S_f , at these temperatures, we conclude that the largest hyperfine field arises from the local moments. In general, however, there can be also an on-site hyperfine interaction to conduction electrons. A more complete description of the hyperfine interactions is given by:

$$\hat{\mathcal{H}}_{\text{hyp}} = \gamma \hbar \hat{\mathbf{I}} \cdot \left(\mathbb{A} \cdot \hat{\mathbf{S}}_c + \sum_{i \in nn} \mathbb{B} \cdot \hat{\mathbf{S}}_f \right), \quad (3)$$

where \mathbb{A} is an on-site hyperfine tensor interaction to the conduction electron spin, and \mathbb{B} is a transferred hyperfine tensor to the Ce 4f spins [11]. A similar scenario is present in

the high temperature superconductors [12]. For the CeMIn_5 materials, the mechanism of the on-site coupling \mathbb{A} is probably due to a combination of core polarization, and polarization of unfilled orbitals with s and p symmetry. The mechanism of the transferred interaction, \mathbb{B} is probably due to a combination of orbital overlap between the 4f wavefunction and ligand nuclei core wavefunctions, and a polarization of the conduction electron spins via the Kondo interaction between $\hat{\mathbf{S}}_c$ and $\hat{\mathbf{S}}_f$. For the remainder of this article, we assume that the hyperfine parameters are material dependent constants, and will not discuss their microscopic mechanism further.

2.1. Coupling to $\text{In}(1)$

In the antiferromagnetic compound CeRhIn_5 ($T_N = 3.8\text{K}$) the magnetic order is an incommensurate spiral with the moments in the ab plane, and the modulation along the c direction, with $Q_{\mathbf{AF}} = (0.5, 0.5, 0.297)$ [13, 14]. In this system, the $\text{In}(1)$ lies in the plane with four nearest neighbor Ce spins, and by symmetry one might expect the transferred hyperfine field would vanish in the ordered state. In fact, the hyperfine field is finite ($H_{\text{hyp}} \approx 1.8\text{kOe}$), and is about five times larger than one would expect from a direct dipolar interaction between localized 4f spins and the In ($I = 9/2$) nuclear spin. The reason the transferred hyperfine does not vanish is that the eigenvectors of the tensor \mathbb{B} do not coincide with the unit cell coordinates, but rather point along the bond directions between the Ce and the $\text{In}(1)$. We can write \mathbb{B} as:

$$\mathbb{B} = \begin{pmatrix} B_{\parallel} & 0 & 0 \\ 0 & B_{\perp} & 0 \\ 0 & 0 & B_c \end{pmatrix} \quad (4)$$

in the bond-coordinate system, where B_{α} corresponds to the transferred hyperfine coupling along the α direction as shown in Fig. 2. In the unit cell (and experimental) reference frame, this tensor becomes

$$\mathbb{B}_{1,3} = \begin{pmatrix} B_0 & B_a & 0 \\ B_a & B_0 & 0 \\ 0 & 0 & B_c \end{pmatrix} \quad (5)$$

and

$$\mathbb{B}_{2,4} = \begin{pmatrix} B_0 & -B_a & 0 \\ -B_a & B_0 & 0 \\ 0 & 0 & B_c \end{pmatrix} \quad (6)$$

for the sites $i \in (1, 2, 3, 4)$ in Fig. 1, where $B_0 = (B_{\parallel} + B_{\perp})/2$ and $B_a = (B_{\parallel} - B_{\perp})/2$.

The on-site interaction, \mathbb{A} is probably anisotropic, but diagonal in the unit cell coordinates:

$$\mathbb{A} = \begin{pmatrix} A_{ab} & 0 & 0 \\ 0 & A_{ab} & 0 \\ 0 & 0 & A_c \end{pmatrix} \quad (7)$$

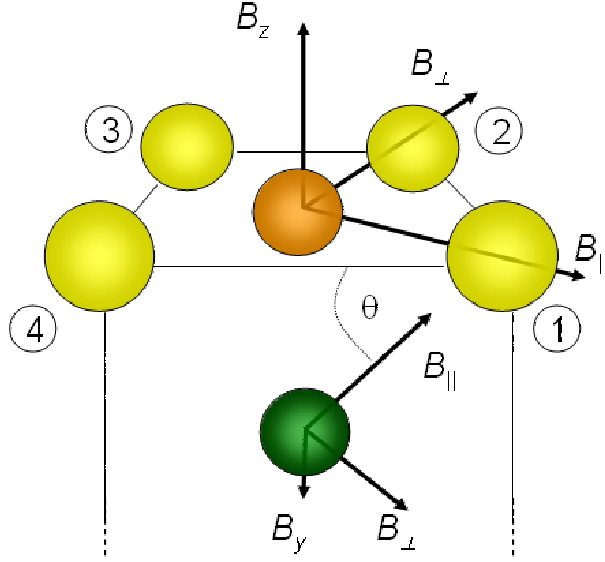


Figure 2. The principle axes of the transferred hyperfine tensors \mathbb{B}_i of the In(1) and the In(2) sites.

The Knight shift at the In(1) site, then should be.

$$\begin{aligned} K_\alpha(T) &= \left(A_\alpha \langle \hat{S}_c^\alpha \rangle + 4B_\alpha \langle \hat{S}_f^\alpha \rangle \right) / H_0 \\ &\approx 4B_0 \chi_\alpha(T), \end{aligned} \quad (8)$$

where the approximation should hold for temperatures $T > T^*$, where the Curie-Weiss susceptibility of the 4f electrons dominates the Pauli susceptibility of the conduction electrons. Below, in section 3, we discuss the Knight shift for $T < T^*$, where the approximation no longer holds.

In the antiferromagnetic state of CeRhIn_5 the ordering is given by:

$$\langle \mathbf{S}_f(\mathbf{r}) \rangle = S_0 \cos\left(\frac{\pi x}{a}\right) \cos\left(\frac{\pi y}{a}\right) (\cos(2\pi q_0 z) \hat{\mathbf{x}} + \sin(2\pi q_0 z) \hat{\mathbf{y}}), \quad (9)$$

where q_0 is the wavevector of the spiral along the c direction [13, 14]. The hyperfine field at the In(1) site is then given by:

$$\mathbf{H}_{\text{hyp}} = 4B_a S_0 (\sin(2\pi q_0 z) \hat{\mathbf{x}} + \cos(2\pi q_0 z) \hat{\mathbf{y}}). \quad (10)$$

$H_{\text{hyp}} \propto (B_{\parallel} - B_{\perp})$ would vanish if \mathbb{B} did not have off-diagonal terms in the coordinates of the unit cell. Also, since $\mathbf{H}_{\text{hyp}} \perp c$, and the electric field gradient (EFG) vector for the In(1) is parallel to c , the resonance frequency in the antiferromagnetic state is independent of z . Consequently, even though the magnetism is incommensurate, the In(1) lines remain sharp [13]. The orientation of the 4f moments and the hyperfine fields are shown in Fig. 3.

A further consequence of anisotropic transferred hyperfine tensor, \mathbb{B} is that the In(1) is sensitive to the critical fluctuations of the order parameter above T_N . We will come back to this subject later in a discussion of the form factors.

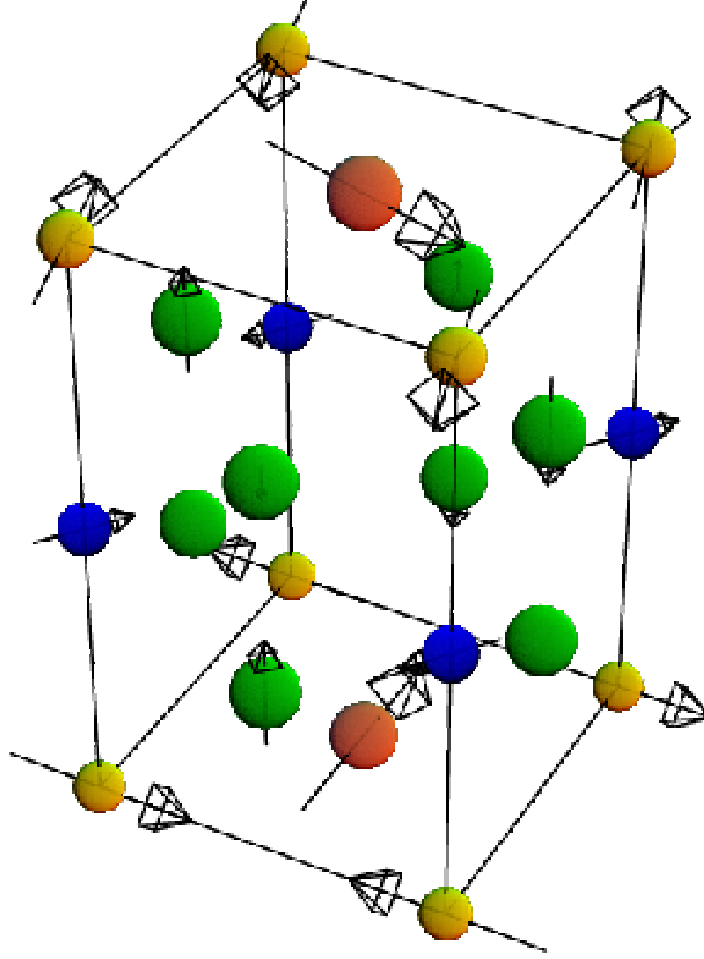


Figure 3. The hyperfine fields in the CeMIn_5 materials. The atoms are the same as in Fig. 1. The arrows on the Ce atoms denote the direction of the electronic moments, for the ordered structure in the CeRhIn_5 antiferromagnetic state. The arrows on the other atoms (In(1), In(2) or M) denote the direction of the hyperfine fields at the particular site, as discussed in the text. An animated version of this figure is available online, showing how the hyperfine fields change as the direction of the Ce moments changes.

2.2. Coupling to In(2)

The In(2) is in a low symmetry site out of the Ce plane. We expect that the transferred hyperfine coupling tensor to this site should also be diagonal along the bond axes (see Fig. 2). In the unit cell coordinate system, we have then:

$$\mathbb{B}_1 = \begin{pmatrix} B_0 + B_a \cos 2\theta & 0 & B_a \sin 2\theta \\ 0 & B_y & 0 \\ B_a \sin 2\theta & 0 & B_0 - B_a \cos 2\theta \end{pmatrix} \quad (11)$$

and

$$\mathbb{B}_2 = \begin{pmatrix} B_0 + B_a \cos 2\theta & 0 & -B_a \sin 2\theta \\ 0 & B_y & 0 \\ -B_a \sin 2\theta & 0 & B_0 - B_a \cos 2\theta \end{pmatrix} \quad (12)$$

where θ is the angle between the Ce-In(2) direction and the a axis. For CeRhIn_5 $\theta \approx 45^\circ$. In this case, the Knight shift is given by:

$$K_a = \left(A_a \langle \hat{S}_c^a \rangle + 2(B_0 + B_a \cos(2\theta)) \langle \hat{S}_f^a \rangle \right) / H_0 \quad (13)$$

$$K_b = \left(A_b \langle \hat{S}_c^b \rangle + 2B_y \langle \hat{S}_f^b \rangle \right) / H_0 \quad (14)$$

$$K_c = \left(A_c \langle \hat{S}_c^c \rangle + 2(B_0 - B_a \cos(2\theta)) \langle \hat{S}_f^c \rangle \right) / H_0, \quad (15)$$

where we have ignored the small component of hyperfine field that is perpendicular to H_0 . As in the case for In(1), we assume that the Pauli susceptibility is negligible for $T > T^*$, so

$$K_a \approx 2(B_0 + B_a \cos(2\theta)) \chi_{ab}(T) \quad (16)$$

$$K_b \approx 2B_y \chi_{ab}(T)$$

$$K_c \approx 2(B_0 - B_a \cos(2\theta)) \chi_c(T).$$

Even though the spin susceptibility is isotropic in the ab plane, the In(2) shift depends on whether the field is in the a or b directions. The reason for this is that the symmetry of the In(2) site is lowered when the field is in the plane, so two of the In(2) sites have the field applied parallel to the face of the unit cell, and two of the sites have a field perpendicular to the face of the unit cell.

In the Neél state, the hyperfine field at the In(2) site is parallel to the c axis:

$$\mathbf{H}_{\text{hyp}} = 2B_a S_0 \cos(2\pi q_0 z) \hat{\mathbf{z}}. \quad (17)$$

Once again, the hyperfine field at the In(2) site is finite only because the tensor \mathbb{B} is not diagonal in the unit cell coordinates, and thus the In(2) is also sensitive to critical fluctuations. The orientation of the hyperfine field at the In(2) sites is shown in Fig. 3. Note that in the case of the In(2), the resonance frequency is dependent on the z coordinate, therefore the In(2) exhibits a broad "powder pattern"-type resonance spectrum in the ordered state of CeRhIn_5 [13].

2.3. Coupling to M

For the M site, the bond axes coincide with the unit cell axes, so the hyperfine tensor is diagonal in the unit cell basis. We thus have:

$$\mathbb{B}_{1,2} = \begin{pmatrix} B_{ab} & 0 & 0 \\ 0 & B_{ab} & 0 \\ 0 & 0 & B_c \end{pmatrix}, \quad (18)$$

and

$$K_{ab} = \left(A_{ab} \langle \hat{S}_c^a \rangle + B_{ab} \langle \hat{S}_f^{ab} \rangle \right) / H_0 \quad (19)$$

$$K_c = \left(A_c \langle \hat{S}_c^c \rangle + B_c \langle \hat{S}_f^c \rangle \right) / H_0, \quad (20)$$

so for $T > T^*$,

$$K_{ab} \approx 2B_{ab}\chi_{ab}(T) \quad (21)$$

$$K_c \approx 2B_c\chi_c(T).$$

In the interest of notational simplicity, we have not included any indices on the on-site and transferred hyperfine coupling constants, A_α and B_α for the different sites (In(1), In(2) and M). In general, each one of these couplings is different, and is material dependent.

3. The Knight Shift Anomaly

In a generic Kondo lattice system, there are two spin species, \hat{S}_c and \hat{S}_f . Therefore, there are three distinct spin susceptibilities: $\chi_{cc} = \langle \hat{S}_c \hat{S}_c \rangle$, $\chi_{cf} = \langle \hat{S}_c \hat{S}_f \rangle$, and $\chi_{ff} = \langle \hat{S}_f \hat{S}_f \rangle$. In Eqs. 8, 16 and 21 we made the approximation that $\langle S_c \rangle \ll \langle S_f \rangle$ for $T \gg T^*$. A more complete description is given by $g_c \mu_B \langle S_c \rangle = (\chi_{cc}(T) + \chi_{cf}(T))H_0$ and $g_f \mu_B \langle S_f \rangle = (\chi_{ff}(T) + \chi_{cf}(T))H_0$, where $g_{c,f}$ are the g-factors. In fact, the full expression for the Knight shift is given by:

$$K_\alpha^{(\eta)}(T) = \left(A_\alpha^{(\eta)} \right) \chi_{cc,\alpha}(T) + \left(A_\alpha^{(\eta)} + \sum_{i \in nn} B_\alpha^{(\eta)} \right) \chi_{cf,\alpha}(T) + \left(\sum_{i \in nn} B_\alpha^{(\eta)} \right) \chi_{ff,\alpha}(T), \quad (22)$$

where η refers the the particular nuclear site, α is the field direction, and for simplicity we have absorbed the g-factors into the definition of the hyperfine constants [11]. The bulk susceptibility is given by:

$$\chi_\alpha(T) = \chi_{cc,\alpha}(T) + 2\chi_{cf,\alpha}(T) + \chi_{ff,\alpha}(T). \quad (23)$$

Note that if $A_\alpha^{(\eta)} = \sum_{i \in nn} B_\alpha^{(\eta)}$, then the relation $K \propto \chi$ is recovered, however in general this relation will not hold. If $\chi_{cc}(T)$, $\chi_{cf}(T)$, and $\chi_{ff}(T)$ have different temperature dependences, then the Knight shift will not be proportional to susceptibility, leading to a Knight shift anomaly at a temperature T^* .

Since a complete solution of the Kondo lattice remains a challenge for theory at present, there is no description of the temperature dependence of $\chi_{cc}(T)$, $\chi_{cf}(T)$, and $\chi_{ff}(T)$. However, using empirical observations Nakatsuji and coworkers developed a phenomenological two-fluid picture of the susceptibility and specific heat in CeCoIn_5 which also works well for understanding the Knight shift anomaly [15, 11, 9]. In this picture, $\chi_{cf}(T) \sim (1 - T/T^*) \log(T^*/T)$ for $T < T^*$, and $\chi_{cf}(T) \sim 0$ for $T > T^*$, and we assume χ_{cc} is negligible for all temperatures. This relation appears to hold for several

different heavy fermion and mixed valent compounds down to the relevant ordering temperatures [11, 16]. Recently we have found evidence that $\chi_{cf}(T)$ saturates below a temperature $T_0 \ll T^*$ in CeCoIn_5 for $H > H_{c2}$. Recent theoretical work lends support for the two-fluid picture [17, 18], but a full description is still lacking. Nevertheless, the NMR data in a wide range of materials strongly suggest that a complete theoretical description should involve one localized degree of freedom giving rise to \hat{S}_f , and another delocalized degree of freedom, giving rise to \hat{S}_c .

4. The Spin Lattice Relaxation Rate and the Form Factors

The spin lattice relaxation of the nuclei in the CeMnIn_5 materials is dominated by magnetic fluctuations mediated by the dynamic hyperfine fields at the nuclear sites. The spin lattice relaxation rate, T_1^{-1} , can be written in general as:

$$\frac{1}{T_{1,\alpha}} = \gamma_n^2 \sum_{\beta} \int_0^{\infty} \langle H_{\beta}(t) H_{\beta}(0) \rangle e^{i\omega_L t} dt, \quad (24)$$

where we have dropped the "hyp" notation for simplicity, ω_L is the Larmor frequency, γ_n is the nuclear gyromagnetic ratio, and the sum is over the two spatial directions orthogonal to the α direction. In general, the hyperfine field is given by:

$$\mathbf{H}(t) = \mathbb{A} \cdot \hat{\mathbf{S}}^c(0, t) + \sum_{i \in \text{nn}} \mathbb{B}^{(i)} \cdot \hat{\mathbf{S}}^f(\mathbf{r}_i, t) \quad (25)$$

where the i -sum is over the nearest neighbor Ce spins, and the γ sum is over the spatial directions. This expression can be simplified by writing:

$$H_{\beta}(t) = \sum_{\mathbf{q}, \gamma} F_{\beta\gamma}^c(\mathbf{q}) S_{\gamma}^c(\mathbf{q}, t) + F_{\beta\gamma}^f(\mathbf{q}) S_{\gamma}^f(\mathbf{q}, t), \quad (26)$$

where $S_{\beta}^{(c,f)}(\mathbf{q}, t) = \sum_{\mathbf{r}} \hat{S}_{\beta}^{(c,f)}(\mathbf{r}, t) e^{-i\mathbf{q} \cdot \mathbf{r}}$, and the form factors $\mathbb{F}^{(c,f)}(\mathbf{q})$ are given by:

$$F_{\beta\gamma}^c(\mathbf{q}) = A_{\beta\beta} \delta_{\beta\gamma} \quad (27)$$

$$F_{\beta\gamma}^f(\mathbf{q}) = \sum_i B_{\beta\gamma}^{(i)} e^{i\mathbf{q} \cdot \mathbf{r}_i}. \quad (28)$$

We then have:

$$\begin{aligned} \langle H_{\beta}(t) H_{\beta}(0) \rangle &= \sum_{\mathbf{q}, \gamma} (F_{\beta\gamma}^c(\mathbf{q}))^2 \langle S_{\gamma}^{(c)}(\mathbf{q}, t) S_{\gamma}^{(c)}(\mathbf{q}, 0) \rangle + \\ &\quad 2 \sum_{\mathbf{q}, \gamma} F_{\beta\gamma}^c(\mathbf{q}) F_{\beta\gamma}^f(\mathbf{q}) \langle S_{\gamma}^{(c)}(\mathbf{q}, t) S_{\gamma}^{(f)}(\mathbf{q}, 0) \rangle + \\ &\quad \sum_{\mathbf{q}, \gamma} (F_{\beta\gamma}^f(\mathbf{q}))^2 \langle S_{\gamma}^{(f)}(\mathbf{q}, t) S_{\gamma}^{(f)}(\mathbf{q}, 0) \rangle. \end{aligned} \quad (29)$$

Here we have assumed that spin correlations at different \mathbf{q} -vectors and directions are uncorrelated: $\langle S_{\beta}^{(c,f)}(\mathbf{q}, t) S_{\gamma}^{(c,f)}(\mathbf{q}', 0) \rangle = \delta_{\mathbf{q}\mathbf{q}'} \delta_{\beta\gamma} \langle S_{\beta}^{(c,f)}(\mathbf{q}, t) S_{\beta}^{(c,f)}(\mathbf{q}, 0) \rangle$, although such an assumption may not be always valid [19]. Using the fluctuation-dissipation theorem, Eq. 24 can be rewritten as:

$$\frac{1}{T_{1,\alpha}} = \frac{1}{T_{1,\alpha}^{cc}} + \frac{2}{T_{1,\alpha}^{cf}} + \frac{1}{T_{1,\alpha}^{ff}}, \quad (30)$$

where:

$$\frac{1}{T_{1,\alpha}^{(\eta\eta')}} = \gamma_n^2 k_B T \lim_{\omega \rightarrow 0} \sum_{\mathbf{q}, \beta} \phi_{\beta}^{\eta\eta'}(\mathbf{q}) \frac{\chi_{\eta\eta'\beta}''(\mathbf{q}, \gamma)}{\hbar\omega}, \quad (31)$$

where $\chi_{\eta\eta'\beta}''(\mathbf{q}, \omega)$ is the dynamical spin susceptibility along the β direction, and

$$\phi_{\beta}^{\eta\eta'}(\mathbf{q}) = \sum_{\gamma} F_{\beta\gamma}^{(\eta)}(\mathbf{q}) F_{\beta\gamma}^{(\eta')}(\mathbf{q}). \quad (32)$$

Clearly, the behavior of the spin lattice relaxation predicted by Eq. 30 can be quite complex, depending on the temperature dependence of the various dynamical spin susceptibilities χ_{cc}'' , χ_{cf}'' and χ_{ff}'' , especially if the temperature dependence of these three quantities differ. At present, there is no clear theoretical model for how these susceptibilities should behave. However, we can make several important and general conclusions. NMR Knight shift studies and theoretical considerations suggest that χ_{cf} is negligible for $T > T^*$ [20]. Therefore, we expect that $T_1^{-1} \approx T_{1,cc}^{-1} + T_{1,ff}^{-1}$ for $T > T^*$. Below T^* , a new contribution, $T_{1,cf}^{-1}$ can become important, and may lead to an anomalous temperature dependence, as observed in CeCoIn₅ for fields applied in the *ab* plane [21].

The filter functions $\phi_{\beta}^{\eta\eta'}(\mathbf{q})$ given in Eq. 32 enhance or suppress the relative importance of spin fluctuations to relax the nuclei at different parts of the Brillouin zone. For the In(1) we have:

$$\begin{aligned} \phi_{a,b}^{ff}(\mathbf{q}) &= 16B_0^2 \cos^2(q_x a/2) \cos^2(q_y a/2) \\ &\quad + 16B_a^2 \sin^2(q_x a/2) \sin^2(q_y a/2) \end{aligned} \quad (33)$$

$$\phi_c^{ff}(\mathbf{q}) = 16B_z^2 \cos^2(q_x a/2) \cos^2(q_y a/2) \quad (34)$$

Note that $F_{\beta}^c(\mathbf{q}) = A_{\beta\beta}$ is \mathbf{q} -independent. For the In(2),

$$\begin{aligned} \phi_a^{ff}(\mathbf{q}) &= 4(B_0 + B_a \cos 2\theta)^2 \cos^2(q_x a/2) + \\ &\quad 4(B_a \sin 2\theta)^2 \sin^2(q_x a/2) \end{aligned} \quad (35)$$

$$\phi_b^{ff}(\mathbf{q}) = 4B_y^2 \cos^2(q_x a/2) \quad (36)$$

$$\begin{aligned} \phi_c^{ff}(\mathbf{q}) &= 4(B_0 - B_a \cos 2\theta)^2 \cos^2(q_x a/2) + \\ &\quad 4(B_a \sin 2\theta)^2 \sin^2(q_x a/2) \end{aligned} \quad (37)$$

and for the M site:

$$\phi_a^{ff}(\mathbf{q}) = 4B_{ab}^2 \cos^2(q_z c/2) \quad (38)$$

$$\phi_c^{ff}(\mathbf{q}) = 4B_c^2 \cos^2(q_z c/2) \quad (39)$$

These quantities are plotted in Figs. 4 and 5. The anisotropic nature of the hyperfine tensor of the In(1) and In(2) gives rise to a second component in their filter functions that does not vanish at $\mathbf{q} = (\pi/a, \pi/a, q_z)$. A priori, one might expect that the In(1) and In(2) would be insensitive to fluctuations at these wavevectors because they are located in symmetric positions in the unit cell with respect to the Ce. However, since the bond axes are not coincident with the unit cell axes, the hyperfine tensor is not diagonal. As

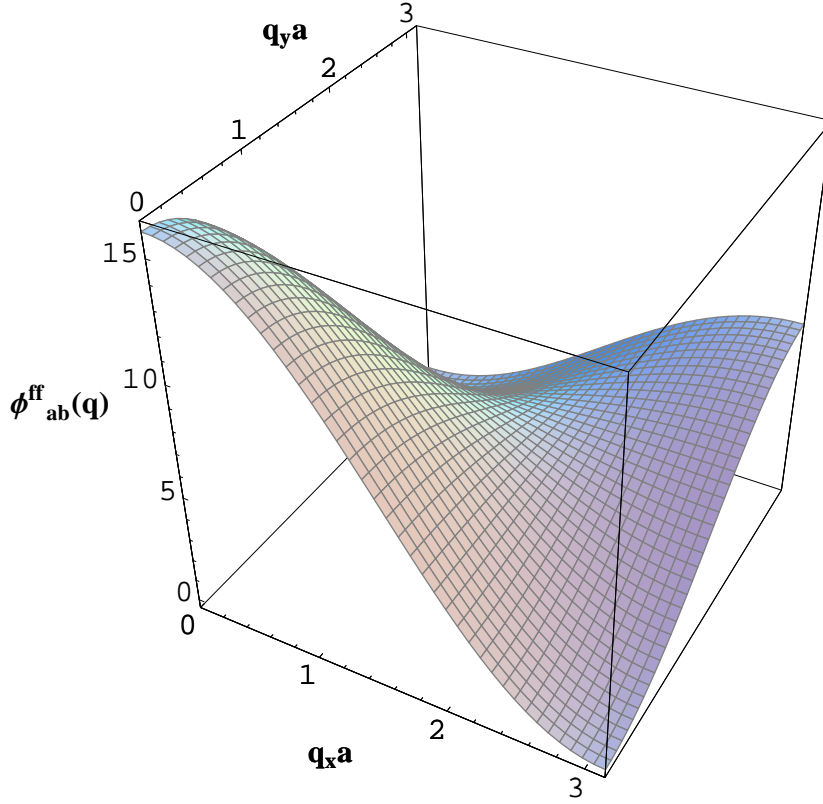


Figure 4. The filter function $\phi_{ab}^{ff}(\mathbf{q})$ for the In(1), shown for $B_a/B_0 = 0.6$. Note that $\phi_{ab}^{ff}(\mathbf{q})$ remains finite for $\mathbf{q} = (\pi/a, \pi/a, q_z)$.

a result, the In(1) and In(2) are sensitive to antiferromagnetic fluctuations [21]. In the cuprates, there is no such anisotropy in the hyperfine tensors, since the bond axes of the Cu and the O nuclei lie along the unit cell directions.

It should be pointed out that the the spin lattice relaxation at the In(2) site is particularly difficult to interpret, because the electric field gradient (EFG) does not have axial symmetry. The quadrupolar part of the hamiltonian of the In(2) is given by:

$$\hat{\mathcal{H}}_Q = \frac{h\nu_{cc}}{6} \left[(3\hat{I}_c^2 - \hat{I}^2) + \frac{\nu_{aa} - \nu_{bb}}{\nu_{cc}} (\hat{I}_a^2 - \hat{I}_b^2) \right] \quad (40)$$

where the $\nu_{\alpha\alpha}$ are the eigenvalues of the EFG tensor [22]. Since $\hat{\mathcal{H}}_Q$ does not commute with \hat{I}_a , \hat{I}_b or \hat{I}_c , the eigenstates of the nuclear levels are non-trivial superpositions of the $|m\rangle$ states, and the usual selection rules for transitions between nuclear levels do not hold. Consequently, unlike the In(1) which does have axial symmetry, the master equation which governs the relaxation of the In(2) nuclei cannot be solved analytically. One can measure the relaxation at the In(2) site, which is a function of the rates given in Eq. 24, but it is not straightforward to independently measure the rates $T_{1,\alpha}^-$.

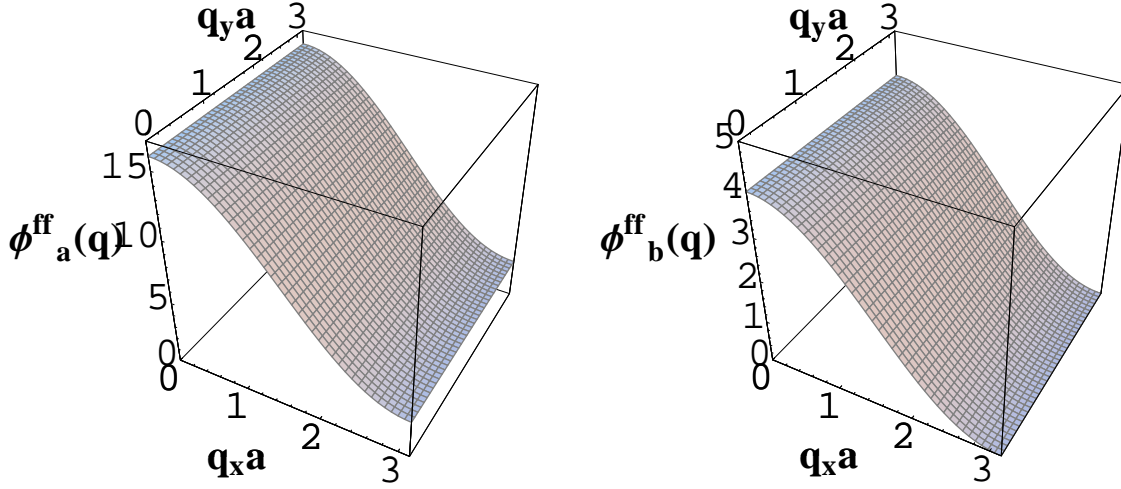


Figure 5. The filter functions $\phi_a^{ff}(\mathbf{q})$ and $\phi_b^{ff}(\mathbf{q})$ for the In(2), shown for $B_a/B_0 = 0.3$. Note that $\phi_a^{ff}(\mathbf{q})$ remains finite for all \mathbf{q} , whereas $\phi_b^{ff}(\mathbf{q})$ does not.

In fact, several groups have measured the temperature dependence of the relaxation rate at the In(1) site in the various 115 compounds, and have found a rich variety of behavior both in the superconducting and normal states [23, 24, 25, 26, 27]. In CeRhIn_5 , the temperature dependence of the spin lattice relaxation is a strong function of pressure [23], and even shows signs reminiscent of the pseudogap observed in the high temperature superconductors. One explanation is that the spin fluctuations are modified with pressure as the ground state evolves from antiferromagnetic to superconducting. However, the internal field in the ordered state is reduced linearly with pressure, while the ordered moment remains constant [28]. This result suggests that the hyperfine constants may change with pressure, a result that has not yet been confirmed by Knight shift measurements under pressure. If the hyperfine constants are pressure dependent, then the temperature dependence of the T_1^{-1} dominantly reflects these changes, rather than changes in the spin fluctuation spectrum. Measurements of T_1^{-1} in $\text{CeRh}_{1-x}\text{Ir}_x\text{In}_5$ alloys and in CeRhIn_5 under pressure suggest that there are regions of the phase diagram where both antiferromagnetism and superconductivity coexist microscopically [23, 27]. In such cases, it appears that the typical T^3 temperature dependence in the superconducting state of d-wave superconductors may be modified, possibly due to the presence of the antiferromagnetic order. Further studies of the Knight shift in these systems may shed important new light on this behavior.

5. NMR in the Actinide 115's

Recently the actinide based 115's, synthesized with U, Np and Pu have attracted attention due to the large superconducting transition temperature in PuCoGa_5 [4, 5].

One of the striking discoveries in $PuCoGa_5$ is that the temperature dependence of spin lattice relaxation rate scales with the transition temperature in the same manner in $CeCoIn_5$, $PuCoGa_5$, and $YBa_2Cu_3O_7$ suggesting a common mechanism. Although there is little detailed information available about the hyperfine couplings present in the $PuMnGa_5$ materials, there is no reason to suspect that the general form of the tensors \mathbb{A} and \mathbb{B} should differ from those in the $CeMn_5$ materials. Therefore, the spin lattice relaxation rate measurements, whether on the In(1), In(2), Ga(1) or Ga(2), should be sensitive to antiferromagnetic fluctuations at $(\pi/a, \pi/a, q_z)$. The unusual temperature dependence of T_1^{-1} may well reflect the temperature dependence of χ''_{ff} and χ''_{cf} . The spin lattice relaxation rate measured in $PuRhGa_5$, however, differs dramatically from that found in $PuCoGa_5$ below $\sim 30K$, well above T_c [29]. Although this anomaly may reflect unusual spin dynamics [30], another explanation may be the presence of a Knight shift anomaly at 30K observed in this material [31]. This discrepancy highlights the unusual hyperfine interactions present in all of the heavy fermion materials, and demonstrates the need for a general understanding of the two spin components and their dynamics before placing too much emphasis on the behavior of the spin lattice relaxation as a function of temperature.

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